Nonlinear coupling of continuous variables at the single quantum level

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(Dated: February 1, 2008)

We experimentally investigate nonlinear couplings between vibrational modes of strings of cold ions stored in linear ion traps. The nonlinearity is caused by the ions' Coulomb interaction and gives rise to a Kerr-type interaction Hamiltonian $H = \hbar\chi\hat{n}_r\hat{n}_s$, where $\hat{n}_r, \hat{n}_s$ are phonon number operators of two interacting vibrational modes. We precisely measure the resulting oscillation frequency shift and observe a collapse and revival of the contrast in a Ramsey experiment. Implications for ion trap experiments aiming at high-fidelity quantum gate operations are discussed.

PACS numbers: 03.67.-a, 05.45.Xt, 32.80.Pj

Kerr nonlinearities in optical media caused by dispersively acting third-order susceptibilities play an important role in nonlinear optics. There is considerable interest in realizing a Hamiltonian $H = \hbar\chi\hat{n}_r\hat{n}_s$ coupling two field modes by a cross-Kerr nonlinearity as this interaction might be used for quantum-nondemolition measurements of single photons [1], for the realization of quantum gate operations in photonic quantum computation [2, 3, 4, 5] and for photonic Bell state detection [6]. Unfortunately, the required strong nonlinearities are difficult to realize experimentally [7, 8].

In trapped ion quantum computing, continuous quantum variables occur in the description of the joint vibrational modes of the ion string. The normal modes are of vital importance for all entangling quantum gates as they can give rise to effective spin-spin couplings in laser-ion interactions [9]. The normal mode picture naturally appears when the ion trap potential is modelled as a harmonic (pseudo-)potential and the mutual Coulomb interaction between the ions is linearized around the ions' equilibrium positions [10]. In this way, the collective ion motion is described by a set of independent harmonic oscillators with characteristic normal mode frequencies. Small deviations from this picture arise because of the nonlinearity of the Coulomb interaction giving rise to a cross-coupling between the normal modes. While nonlinear Coulomb couplings between ions found lots of attention in ion trap experiments investigating order-chaos transitions [11, 12], its effect in the normal mode regime has mostly gone unnoticed. Here, we are not interested in resonant mode coupling [13] but rather in dispersive cross-Kerr effects leading to shifts of the normal mode frequencies. In this paper, we show that the Coulomb nonlinearity gives rise to a Kerr-type Hamiltonian and we quantitatively measure the strength of the induced frequency shift. This frequency shift is of importance for ion trap experiments aiming at pushing the fidelity of entangling quantum gates towards the fault-tolerant threshold.

For two cold ions of mass m and charge e held in an anisotropic harmonic potential characterized by trap frequencies $\omega_z, \omega_x = \omega_y \equiv \omega_\perp$, with $\omega_z < \omega_\perp$, the equilibrium positions of the ions are given by $\mathbf{r_i} = (0, 0, \pm z_0)$, where $z_0 = (e^2/(16\pi\epsilon_0 m\omega_z^2))^{1/3}$. Separating the center-of-mass (COM) and the relative ion motion by introducing the coordinates $\mathbf{R} = (\mathbf{r}_2 + \mathbf{r}_1)/2$ and $\mathbf{r} = (\mathbf{r}_2 - \mathbf{r}_1)/2$ yields the potential energy

$$U_{pot} = U_{pot}^{COM}(\mathbf{R}) + m{\omega_z}^2\Big(z^2 + \frac{\omega_\perp^2}{\omega_z^2}(x^2+y^2) + \frac{2z_0^3}{r}\Big).$$

Expanding the Coulomb potential to fourth order around $(0, 0, z_0)$ by setting $z = z_0 + u$ and keeping only those terms giving rise to a cross-coupling between normal modes, we find

$$U_{pot}^{rel} = m{\omega_s}^2u^2 + m{\omega_r}^2(x^2+y^2) + V^{(3)} + V^{(4)},$$

where $\omega_s = \sqrt{3}\,\omega_z$, $\omega_r = \sqrt{\omega_\perp^2 - \omega_z^2}$ are the stretch and the rocking mode frequencies [14] and where $V^{(3)}$,$V^{(4)}$ defined by

$$V^{(3)} = \frac{m{\omega_s}^2}{z_0}u(x^2+y^2),\ V^{(4)} = -\frac{2m{\omega_s}^2}{z_0^2}u^2(x^2+y^2)$$

describe the cubic and quartic nonlinearities. Next, we use perturbation theory for a calculation of the energy shifts $\epsilon(n_s, n_r^x, n_r^y)$ of the quantum states described by the quantum numbers (n_s, n_r^x, n_r^y) where n_s and $n_r^{x,y}$ specify the number of stretch mode and rocking mode phonons. In first-order, we calculate only the shift induced by the quartic term as the cubic term does not contribute. In second order, the quartic term can be neglected. The shift of the stretch mode frequency by the rocking mode quantum numbers is given by

$$\delta\omega_s = \frac{\epsilon(n_s+1, n_r^x, n_r^y) - \epsilon(n_s, n_r^x, n_r^y)}{\hbar} = \chi(n_r^x + n_r^y + 1)$$

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with the cross-Kerr coupling constant

$$\chi = -\omega_s \left(1 + \frac{\omega_s^2/2}{4\omega_r^2 - \omega_s^2}\right)\left(\frac{\omega_z}{\omega_r}\right)\left(\frac{2\hbar\omega_z}{\alpha^2 mc^2}\right)^{\frac{1}{3}}, \qquad (1)$$

where α denotes the fine structure constant and c the speed of light. This expansion is valid as long as $\omega_r/\omega_s \gg (x_0/z_0)$ and $|2\omega_r - \omega_s|/\omega_s \gg (u_0/z_0)$ where x_0 (u_0) are the spatial half width of the rocking (stretch) mode ground states, respectively. The resonance $2\omega_r = \omega_s$ corresponds to a parametric coupling converting two rocking mode phonons into a single stretch mode excitation. In experiments with thermally occupied rocking modes, the cross-mode coupling causes dephasing of the stretch mode motion.

In our experiments, two $^{40}Ca^+$ ions are confined in a linear Paul trap with radial trap frequencies of about $\omega_\perp/2\pi = 4$ MHz. By varying the trap's tip voltages from 500 to 2000 V, the axial center-of-mass frequency ω_z is changed from 860 kHz to 1720 kHz. The ions are Doppler-cooled on the $S_{1/2} \leftrightarrow P_{1/2}$ transition. Sideband cooling on the $S_{1/2} \leftrightarrow D_{5/2}$ quadrupole transition [15] prepares the stretch mode in the motional ground state $|0\rangle_s$. Simultaneous cooling of stretch and rocking modes is accomplished by alternating the frequency of the cooling laser exciting the quadrupole transition between the different red motional sidebands. Motional quantum states are coherently coupled by a laser pulse sequence exciting a single ion on the $|S\rangle \equiv S_{1/2}(m = -1/2) \leftrightarrow |D\rangle \equiv D_{5/2}(m = -1/2)$ transition with a focused laser beam on the carrier and the blue sideband. Internal state superpositions $(|S\rangle + e^{i\phi}|D\rangle)|0\rangle$ can be mapped to motional superpositions $|D\rangle(|0\rangle + e^{i\phi}|1\rangle)$ by a π pulse on the blue motional sideband and vice versa. We discriminate between the quantum states $S_{1/2}$ and $D_{5/2}$ by scattering light on the $S_{1/2} \leftrightarrow P_{1/2}$ dipole transition and detecting the presence or absence of resonance fluorescence of the individual ions with a CCD-camera. A more detailed account of the experimental setup is given in Ref. [16, 17].

For a measurement of the stretch mode coherence, a Ramsey experiment between motional states is performed by a laser interacting with only one of the ions, the second ion being just a spectator that modifies the normal mode structure. Starting from the state $|S\rangle|0\rangle$, the superposition state $|D\rangle(|0\rangle + |1\rangle)$ is created by a $\pi/2$ pulse on the carrier transition followed by a π pulse on the blue sideband. During a waiting time of duration τ, the state evolves into $|D\rangle(|0\rangle + e^{i\phi}|1\rangle)$, where ϕ is a random variable. Finally, the motional superposition is mapped back to superposition of internal states $(|S\rangle + e^{i\phi}|D\rangle)|0\rangle$ and probed by a $\pi/2$ pulse on the carrier transition followed by a quantum state measurement. We measure the coherence by varying the phase of the last $\pi/2$ pulse from 0 to 2π and measuring the contrast $C(\tau)$ of the resulting Ramsey pattern as a function of τ. The measurement shown in Fig. 1 exhibits a strong reduction in contrast after only 2 ms. This fast decay could be attributed neither to motional heating (because the stretch mode heating rate was less than 0.02 phonons/ms), nor to an instability of the trapping potential (since a coherence time of about 50 ms was measured on the center-of-mass mode). To probe the dynamics of the dephasing mechanism, a spin echo experiment was performed on a motional superposition $|0\rangle + |1\rangle$. For this, the populations of the $|0\rangle$ and $|1\rangle$ states were exchanged in the middle of the experiment by a carrier π-pulse sandwiched between two blue sideband π-pulses. As can be seen in Fig. 2,

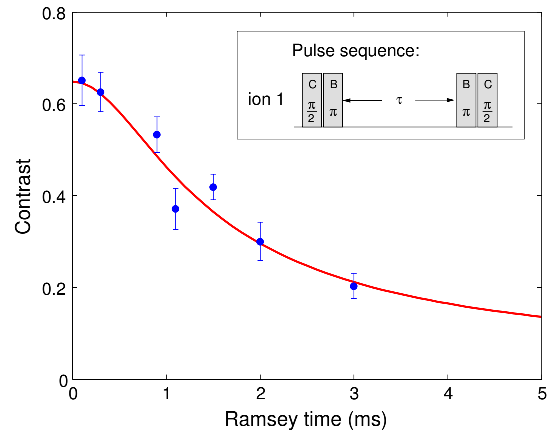

FIG. 1: Contrast $C(\tau)$ of a Ramsey experiment on the stretch mode as a function of the Ramsey waiting time τ. The axial trap frequency was $\omega_z = (2\pi)1486$ kHz, the radial modes were cooled close to the Doppler limit. The initial contrast is limited by imperfect sideband cooling, spurious excitation of the second ion by the focussed laser beam and magnetic field noise. The subsequent loss of contrast is caused by a dephasing of the stretch mode oscillation. The inset shows the pulse sequence of the Ramsey experiment with C denoting carrier pulses and B blue sideband pulses.

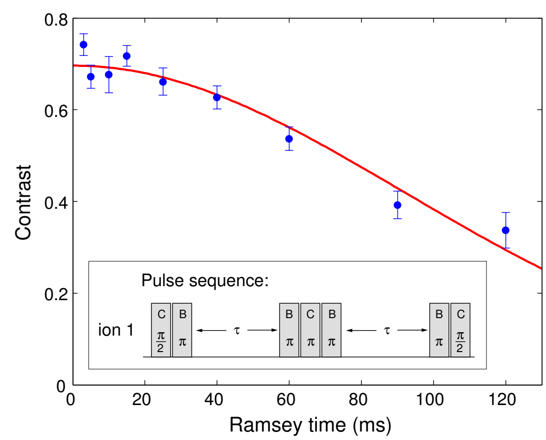

FIG. 2: Contrast $C(\tau)$ of a spin echo experiment on the stretch mode as a function of the Ramsey waiting time τ. Insertion of the spin echo increased the coherence time by nearly two orders of magnitude as compared with a simple Ramsey experiment.

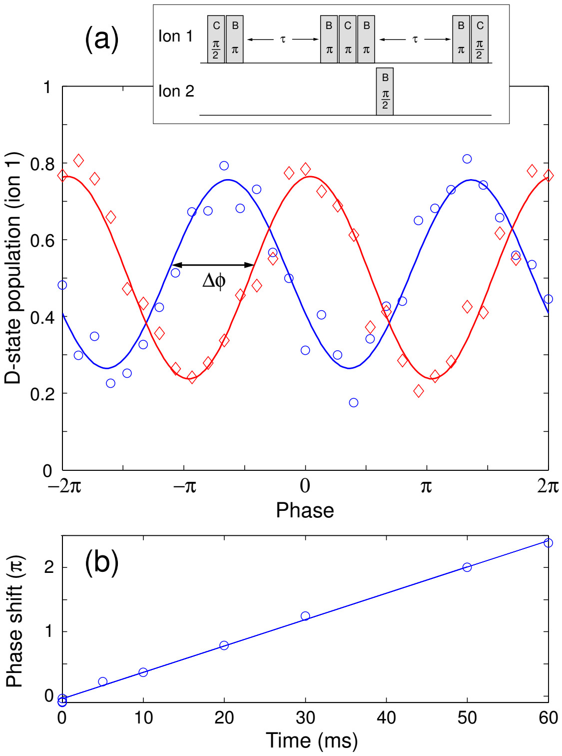

FIG. 3: (a) Shift $\Delta\phi$ of the spin echo phase pattern by a rocking mode phonon observed by measuring the D state population of ion 1 as function of the phase of the last spin echo pulse. The symbol ($\diamond$) labels experiments with an extra phonon in the second spin echo time, ($\circ$) experiments with no extra phonon. (b) Single phonon shift of the spin echo phase pattern as a function of the waiting time. The frequency shift caused by a single rocking mode phonon is inferred from the slope of the function $\Delta\phi(\tau)$.

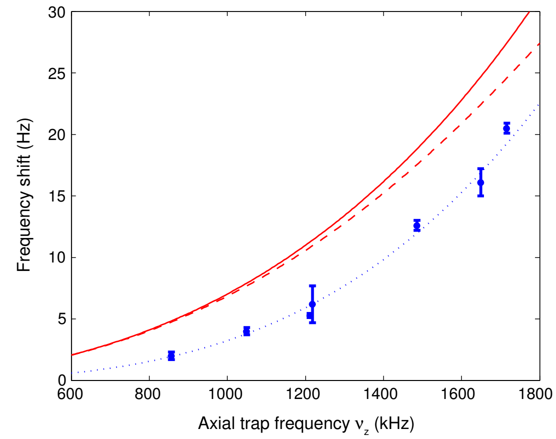

FIG. 4: Shift of the stretch mode frequency pattern by a single rocking mode phonon as a function of the axial trap frequency ω_z. The solid line is the frequency shift predicted by eq. (1), the dashed line shows only the quartic contribution, and the dotted line is a fit to the data using a heuristic scaling law $d\nu_s/dn_r \propto \omega_z^\beta$ with $\beta = 3.25$.

now it takes the spin echo contrast $C(\tau)$ about 100 ms to decay to 50% of its initial value - a clear indication that the stretch mode frequency is fairly stable over the duration of a single experiment but randomly changing from experiment to experiment. This behaviour is expected for a dephasing caused by a thermally distributed rocking mode phonon number that takes on random values at the start of each experiment. To confirm this dephasing mechanism, we remeasured the Ramsey contrast $C(\tau)$ with both rocking modes cooled close to the ground state and observed an increase of the coherence time by a factor of 20 as compared to the experiment shown in Fig. 1.

Having established the cross-coupling between the stretch mode and the rocking mode as the dephasing mechanism, we are interested in quantifying the cross-coupling strength by measuring the stretch mode frequency shift caused by a single rocking mode phonon. In principle, such a measurement could be performed by preparing the rocking modes in a $n_r = 0$ or $n_r = 1$ Fock state and subsequently measuring the stretch mode oscillation frequency in a Ramsey experiment. This, however, would require ultra-stable high-voltage sources for keeping the axial trap frequency ω_z stable to within $10^{-6}\,\omega_z$ or less. To make the experiment more robust against technical noise, we carried out a spin echo experiment instead, where the other ion was excited on the blue sideband of one of the rocking modes directly before the start of the second spin echo waiting time τ (see inset of Fig. 3(a)). If this pulse excites the second ion into the $D_{5/2}$ state, there is exactly one additional phonon in the rocking mode during the second half of the spin echo whose frequency shift is not compensated by the echo sequence. We adjust the duration of this blue sideband pulse so that the excitation is successful in 50% of the experiments and sort the spin echo data into two classes according to the quantum state of the second ion at the end of the experiment. By measuring the phase shift $\Delta\phi(\tau)$ between the two data sets, we are able to infer the shift caused by a single rocking mode phonon. This procedure does not even require cooling the rocking modes to the ground state. Fig. 3(a) shows the resulting phase shift for a waiting time $\tau = 30$ ms. For a calculation of the single phonon frequency shift $d\nu_s/dn_r$, we plot the phase shift as a function of the spin echo time τ, fit a straight line to the data and determine its slope. For the data measured at $\omega_z/2\pi = 1716$ kHz shown in Fig. 3(b), the frequency shift is $d\nu_s/dn_r = 20.5$ Hz/phonon. This measurement procedure was carried out for different axial center-of-mass voltages ω_z while keeping the transverse oscillation frequency $\omega_\perp$ fixed. In Fig. 4 the frequency shift is plotted as a function of ω_z. The experimentally measured frequency shift is slightly smaller than the shift predicted by perturbation theory. The disagreement between experimental data and theoretical model is cur-

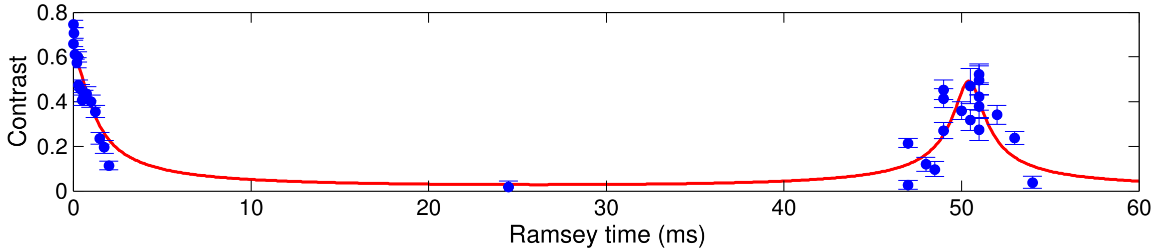

FIG. 5: Collapse and revival of the contrast in a Ramsey experiment probing the motional coherence of the two lowest stretch mode quantum states. The revival occurs at the time predicted by the spin echo experiment for a frequency $\omega_z/2\pi = 1716$ kHz.

rently not understood. It cannot be attributed to the dynamical nature of the trap potential. Using a refined pseudopotential [18] leads only to marginal corrections.

For a thermally occupied rocking mode, the change induced in ω_s is always an integer multiple of the single phonon shift. Therefore, after the initial collapse of the Ramsey contrast (Fig. 1) a revival [19] is to be expected for a time $\tau^* = 2\pi/\chi = 2\pi(d\omega_s/dn_r)^{-1}$. Operating the trap at $\omega_z/2\pi = 1716$ kHz, we indeed observed a revival at the predicted time τ^*. In this experiment, one of the rocking modes was cooled to the ground state while the other one was prepared in a thermal state with $\bar{n}_r \approx 9$ (1σ-confidence interval {5,17}). Fig. 5 shows a fit to the data using the function

$$\tilde{C}(\tau) = e^{-\gamma\tau}|\langle e^{i\chi\hat{n}_r\tau}\rangle| = e^{-\gamma\tau}|\bar{n}_r + 1 - \bar{n}_r e^{i\chi\tau}|^{-1},$$

assuming a thermally occupied rocking mode and an overall loss of contrast $\propto e^{-\gamma\tau}$ accounting for technical noise and motional heating. A fit to the data yields a revival time $\tau^* = 50.5(5)$ ms, an average vibrational quantum number $\bar{n}_r = 9\,(2)$ that is consistent with an independent measurement of $\bar{n}_r$ and a decay rate $\gamma = 0.004(3)\mathrm{s}^{-1}$. The existence of revivals is a further proof of the quantized nature of the rocking motion. The small loss of contrast shows that the probability of a change in the rocking mode phonon number within the interval $[0, \tau^*]$ is quite low.

The observed frequency shifts need to be taken into account in quantum gate realizations operating on the stretch mode. For the parameters [20] used in ref. [9], eq. (1) predicts shifts as big as 100 Hz/phonon, giving rise to a loss of fidelity of about 0.1% for $\bar{n} = 1$.

In summary, we have investigated a nonlinear quantum effect giving rise to a cross-coupling of harmonic oscillators that can be described by a Kerr-like Hamiltonian $H \propto \hat{n}_s\hat{n}_r$. In a two-ion crystal, the nonlinearities lead to a dephasing of the relative ion motion that manifests itself as a collapse of the Ramsey contrast that revives once all oscillations get in phase again. While the nonlinearity is fairly small for the trap parameters investigated here, it could be made bigger by tuning the normal mode frequencies closer to the resonance $\omega_r = 2\omega_s$ so that it might be used for creating entangled motional states of these oscillators. On the other hand, for quantum computing experiments aiming at fault-tolerant quantum gates, the observed nonlinearity points to the necessity of cooling all spectator modes to the ground state or working with transversally very stiff ion traps.

We acknowledge support by the Austrian Science Fund (FWF), the European Commission (SCALA, CONQUEST networks), the US Army Research Office, NSERC and by the Institut für Quanteninformation GmbH. C. R. thanks D. Leibfried for useful discussions and F. Dubin for a critical reading of the manuscript.

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